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Flexible Pressure Sensor with High Sensitivity and Wide Linear Range Based on Janus Gradient Concentration Structure Dual-Sensitive Layers

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Abstract: Flexible pressure sensors, as the core components of wearable electronic devices, the contradiction between their range and sensitivity remains a key bottleneck restricting development. However, developing flexible pressure sensors with both high sensitivity and a wide linear range poses a significant challenge. This study innovatively proposes a Janus dual-sensitive layer structure, constructing a biomimetic pyramid microstructure sensing layer using CNT/CNF composite materials, combined with a PDMS stiffness adjustment layer to achieve mechanical performance regulation. The flexible pressure sensor demonstrates outstanding sensitivity of 18.4 kPa^{-1} and excellent linearity of 99.94% within a broad range of 700 kPa. This design strategy can be extended to other flexible sensor fields that require high sensitivity and a wide linear range, and it is of great significance for the development of high-performance flexible pressure sensors for applications in intelligent robots and wearable devices.

Keywords: Flexible Pressure Sensor, Janus Dual-Sensitive Layer, Carbon Nanotubes, High Sensitivity

1. Introduction

Flexible pressure sensors have a broad application prospect in health monitoring,[1] smart wearables,[2] human-computer interaction,[3]and other fields. The core performance indicators, sensitivity and response range, collectively determine the applicability and accuracy of the sensor in complex scenarios. However, most flexible piezoresistive sensors currently face the issue of a narrow linear range or limited detection range while achieving high sensitivity.

In recent years, researchers have significantly improved the performance of flexible pressure sensors by introducing microstructure design, multilayer sensitive layer structures, and new material systems. [4] For instance, design strategies based on three-dimensional porous foam structures and gradient microstructures have,[5] to some extent, expanded the linear response range of the sensors, but often at the expense of sensitivity. Moreover, these complex design schemes typically require high-precision manufacturing techniques, resulting in a significant increase in production costs and are not conducive to the repeatability and reliability of the sensors.

To overcome the mentioned challenges, this study proposes a Janus gradient conductive structure based on a "double-layer resistance-sensitive structure," achieving an ultra-wide linear response in pressure sensing. By constructing a composite sensitive structure consisting of a high-resistance layer

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and a low-resistance layer on a flexible substrate, a phased switching of current transmission paths within different pressure ranges is realized. This structural design not only effectively alleviates the saturation trend of sensor resistance changes but also significantly extends its linear response range. Furthermore, this study utilizes cellulose nanofibers (CNF) as an additive, combined with carbon nanotubes (CNT) as the sensitive material, to fabricate high-performance flexible pressure sensors. The sensor exhibits a sensitivity of up to 18.4 kPa^{-1} and a linearity of 99.94% within the range of 0~700 kPa. This composite structure based on CNF and CNT not only simplifies the fabrication process and reduces costs but also explores the potential applications of flexible pressure sensors and their extended arrays in the fields of human health and motion monitoring, including gait analysis for posture correction assistance, as well as assessment and rehabilitation for gait instability.

2. Results and discussion

2.1. Design process, structure, and mechanism of flexible pressure sensors

Nanomaterials, with their unique nanoscale size effects and outstanding physical and chemical properties such as ultra-high conductivity, excellent mechanical strength, flexibility, and chemical stability, have shown significant advantages in electronics, sensing, medical fields, and more. Carbon nanotubes (CNTs) are hollow tubular nanomaterials formed by rolling up one or more layers of graphene sheets. They possess ultra-high conductivity and mechanical strength, and their unique piezoresistive effect causes significant changes in the conductive network under external forces, making them ideal sensitive materials for flexible pressure sensors. Cellulose nanofibers (CNFs) are nanoscale fibers extracted from natural cellulose, possessing both excellent flexibility and high mechanical strength. Their hydrophilicity and biocompatibility allow them to disperse easily in aqueous solutions and form stable composite structures with polymers or conductive materials such as CNTs. CNFs not only enhance the flexibility and mechanical stability of sensors but are also suitable for wearable and medical devices due to their natural origin and biodegradability. Polydimethylsiloxane (PDMS) is an organosilicon polymer known for its extraordinary flexibility, chemical inertness, and biocompatibility. It can be stretched to several times its original length without breaking and withstands extreme environments such as acids, bases, and ultraviolet light. When used as a coating layer for sensors, PDMS can adjust stiffness to accommodate different pressure ranges and protect the internal sensitive layer from external environmental interference. This study extensively used nanoscale materials, as shown in Figure 1a, by incorporating CNFs into a polyurethane (PU) substrate, dispersing them evenly by stirring, and utilizing their unique nanostructure and excellent mechanical properties to improve the flexibility and stability of the film. The addition of CNFs not only helps to enhance the mechanical properties of the film but also, to some extent, increases its conductivity. Subsequently, carbon nanotubes (CNTs) were added as sensitive materials to further improve the film's conductivity and mechanical strength. After adding CNTs, stirring continued to ensure thorough and uniform mixing of the materials. Using microelectromechanical systems (MEMS) processes, pyramid structures were etched onto a silicon wafer, and the solution was poured onto the pyramid microstructures to form the desired microstructures. MEMS processes allow for high-precision processing of the film and precise control of the microstructures. The solution was then freeze-dried to remove the moisture from the film while maintaining the integrity of its microstructure. Freeze-drying treatment yields a film with good mechanical and electrical properties. PDMS was used as a stiffness regulator for the film, leveraging its excellent flexibility to provide force dispersion and additional protection and support for the film. The PDMS coating not only prevents the film from being affected by the external environment during preparation and use but also enhances its overall mechanical stability. The flexible pressure sensor

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film prepared through these steps has excellent electrical conductivity, mechanical properties, and flexibility, making it suitable for a variety of applications.

The pyramid V-shaped double-layer resistive sensitive structure is a significant feature of this study, as shown in Figure 1b [6], which illustrates the structural design of our sensor. It consists of a double-layer pyramid-shaped sensing layer made of CNT and CNF, and PDMS as a stiffness regulator. The sensing layer is composed of a composite material made of CNT and CNF, which is the core part of the sensor. CNT has excellent electrical conductivity and mechanical strength, while CNF provides good flexibility and mechanical stability. By mixing CNT and CNF to prepare a thin film, the sensitivity and linear range of the sensor can be significantly improved. The sensing layer is nested within the stiffness regulator, with PDMS as the stiffness layer, wrapping around the sensitive layer externally, serving to protect and adjust the force exerted on the sensor, achieving the regulation of the force exerted on the sensitive layer under different forces. PDMS has good flexibility and elasticity, which can adjust the mechanical properties of the sensor to some extent, allowing it to maintain a good linear response within different pressure ranges. Through encapsulation with polyimide (PI) film and copper electrodes, a flexible pressure sensor is thus fabricated.

The Janus conductive structure achieves a staged switching of current transmission paths within different pressure ranges by constructing a composite sensitive structure of high-resistance and low-resistance layers on a flexible substrate [7]. Its mechanism is shown in Figure 1c, where the left diagram in the text presents a single sensitive layer structure pressure sensor. Due to the limited change in the contact interface area between the electrode and the sensitive layer, the contact resistance changes little, causing the resistance response to saturate quickly under small pressures, with high sensitivity limited to a low-pressure range and decreasing significantly with increasing pressure, resulting in a narrow pressure detection range and poor linearity. The right diagram shows a double sensitive layer structure pressure sensor, which has two contact interfaces, and the current transmission path switches between the two sensitive layers with changes in pressure. These two factors work together to give the sensor with a Janus conductive sensitive layer a wider range of resistance change and slow down the speed at which the resistance response reaches saturation. Therefore, the sensor exhibits a wider pressure sensing range and linearity, and high sensitivity can be maintained over a wider pressure range.

When the sensor is subjected to pressure, on the one hand, the PDMS stiffness regulator disperses a portion of the force, while on the other hand, the remaining force acts on the double-layer sensitive layer with a porous structure, achieving a high sensitivity and wide linear range response of the sensor.

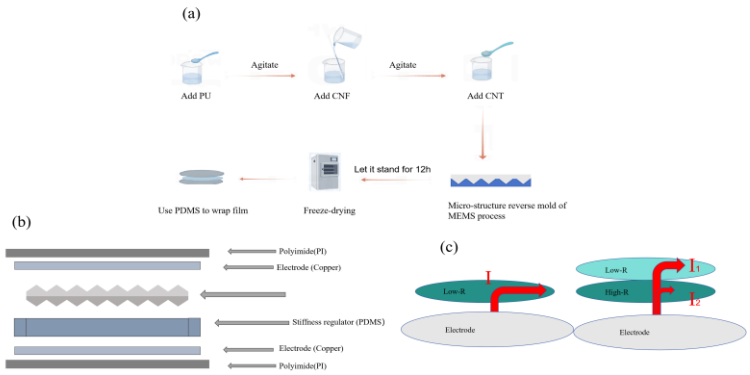

Figure 1: a) Design flowchart of flexible pressure sensors; b) Pyramid V-shaped double-layer resistance sensitive structure diagram; c) Janus gradient concentration structural mechanism diagram

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2.2. The sensing performance of flexible pressure sensors

As shown in Figure 2a, this is the stress-strain curve of the sensor. As the figure shown, the tensile strength of the thin film is 18 MPa, and the elongation at break is 353.33%. Due to the excellent mechanical properties of CNTs, they can act as reinforcement phases. Their nanoscale size effect enables them to effectively disperse stress and prevent the propagation of cracks. When subjected to external forces, CNTs can evenly distribute the stress throughout the material, thereby enhancing the material's tensile strength. CNFs have good flexibility and a high specific surface area, which allows for a good interfacial bond with CNTs. The addition of CNFs can improve the toughness of the composite material. Their fibrous structure can disperse stress through the orientation and entanglement of fibers when the material is stretched, thereby increasing the material's elongation at break.

CNT and CNF form an interwoven network structure within the composite material. The high strength and high modulus of CNT provide good mechanical support for the composite, while the flexibility and dispersibility of CNF contribute to enhancing the overall toughness and ductility of the material. This synergistic effect allows the composite material to better disperse stress and reduce stress concentration points when subjected to external forces, thereby improving tensile strength and elongation at break. Therefore, the high strength and high modulus of CNT, the flexibility and dispersibility of CNF, and the synergistic interaction between the two, collectively enhance the tensile strength and elongation at break of the composite film.

As shown in Figure 2b, this is the sensitivity curve of the sensor. As the figure shown, it can be observed that as the applied pressure increases, the current change rate ($\Delta I/I_0$) exhibits a clear linear growth trend. Specifically, when the pressure increases from 50 kPa to 700 kPa, the current change rate significantly increases from 991.8 to 12961.8. The sensitivity calculated using linear regression is 18.4 kPa-1, indicating that the sensor has high sensitivity and good linear response characteristics. The synergistic effect of CNTs and CNFs not only enhances the mechanical properties of the film but also optimizes its electrical response characteristics, allowing the sensor to demonstrate good current variation at different pressure stages.

As shown in Figure 2c, this is the curve figure of the sensor's recovery response time. As the figure shown, the beginning of pressure application, the current variation rate rapidly increased from 0 to 59, then rapidly rose to 513 between 220 milliseconds and 280 milliseconds, with a response time of 60 milliseconds, indicating that the sensor responds very quickly to pressure changes and can produce significant current responses in a short time. However, after pressure release, the current variation rate gradually decreased to 0 between 480 milliseconds and 550 milliseconds, with a recovery time of 70 milliseconds.

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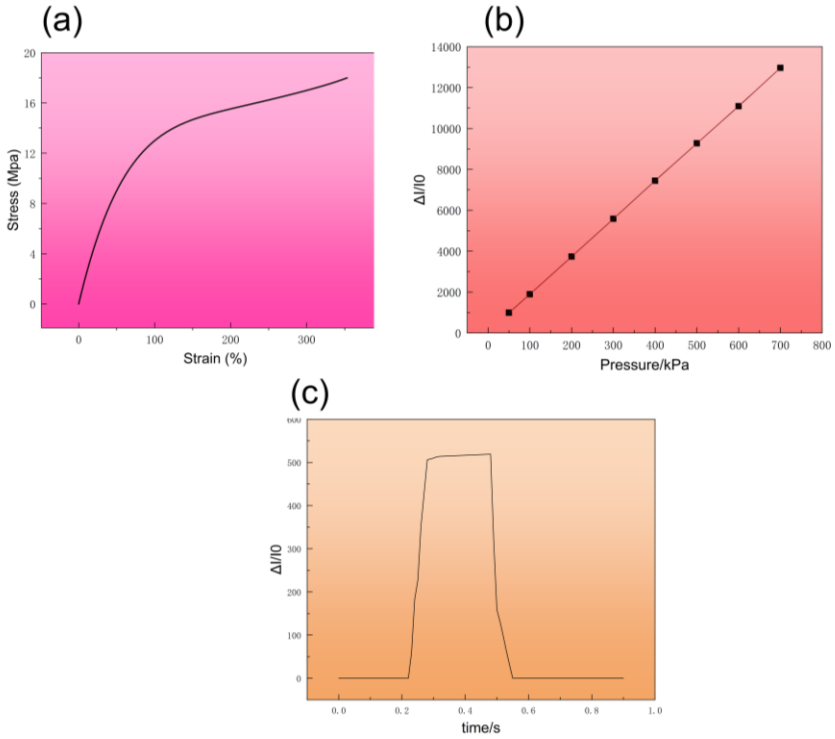

Figure 2: a) Stress-strain curve diagram; b) Curve of sensitivity; c) Response time curve chart

As shown in Figure 3a, this is the curve figure of the sensor under the action of heavy and light objects. The sensor exhibits significantly different current variation characteristics when detecting light and heavy objects. Specifically, from 0 to 5 seconds, the current variation rate ($\Delta I/I_0$) of the sensor is almost 0, indicating that during this stage, the sensor did not detect any significant pressure changes, corresponding to a no-load state. At 10 seconds, a light object was placed on top of a heavy object, and a significant change was observed, indicating that the pressure sensor can respond to both heavier and lighter objects. During the period from 6 to 20 seconds, the current variation rate rapidly increased and stabilized at a higher level (approximately 7300 to 7400), indicating that the sensor detected a significant pressure change during this stage, corresponding to the loading of the heavy object. The sensor exhibits different response characteristics when loading light and heavy objects. When loading light objects, the sensor's current variation is smaller, while when loading heavy objects, the sensor can respond quickly and produce significant current changes.

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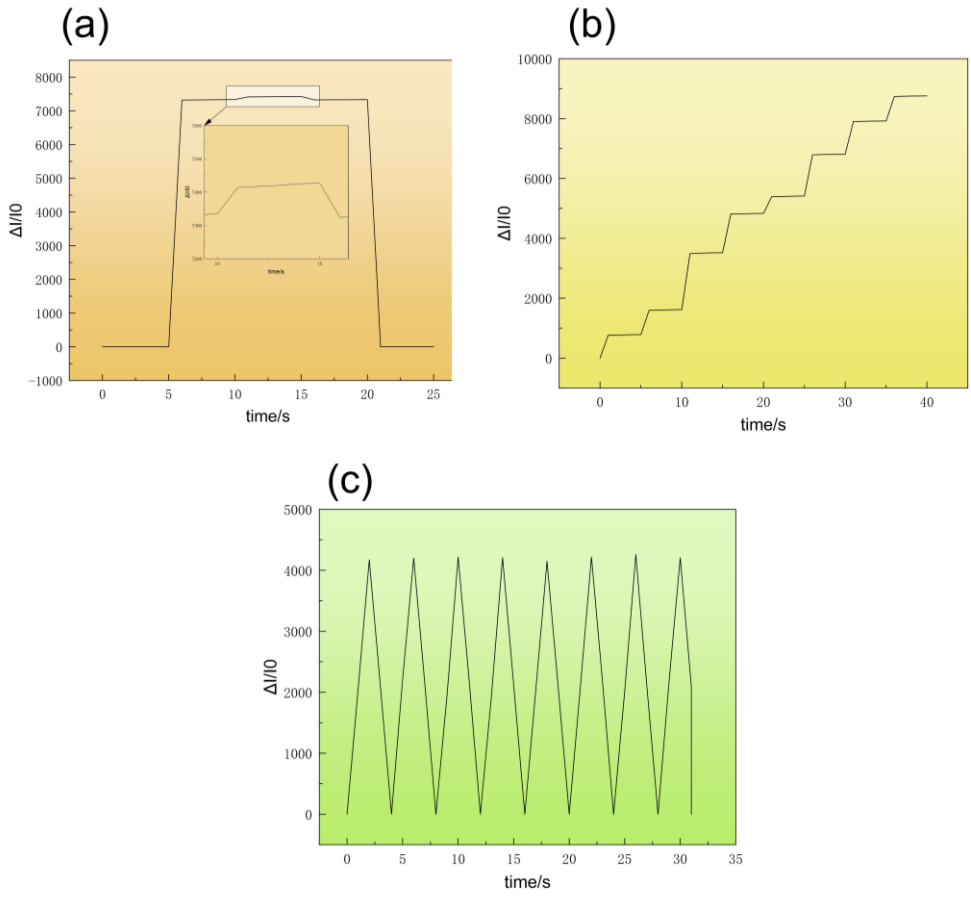

Figure 3: a) Curve figure under the action of heavy and light objects; b) The curve figure of gradually increasing pressure; c) Cyclic curve figure

As shown in Figure 3b, this is the curve figure of the sensor's pressure gradually increasing. As the figure shown, the current variation rate ($\Delta I/I_0$) exhibits a clear stage-wise growth as time increases. In flexible pressure sensors, CNTs, as the sensitive material, their unique nanostructure and excellent electrical properties are key factors affecting current variation. CNTs have a high specific surface area and high conductivity, which can form an efficient conductive network in the film. When pressure is applied, the contact points between CNTs increase, and the conductive paths multiply, resulting in a significant increase in current. Additionally, CNF, as an additive, its flexibility and dispersibility can enhance the uniform distribution of CNTs in the film, further optimizing the stability of the conductive network. The presence of CNF can also improve the mechanical properties of the film, allowing it to better maintain structural integrity when under pressure, thus ensuring the stability and repeatability of current variation.

As shown in Figure 3c, this is the cyclic curve figure of the sensor, which exhibits relatively stable current variation characteristics during multiple cycles of pressure loading and unloading. Specifically, the sensor can respond quickly to pressure changes in each cycle and recover to a current level close to the initial state after the pressure is released. For example, in the first cycle (from 1 second to 4 seconds), the current variation rate ($\Delta I/I_0$) rapidly increased from 0 to 2062 and then quickly dropped back to 0 upon pressure release. In subsequent cycles, the sensor's current variation rate consistently reached a high value upon loading pressure and quickly returned to 0 upon pressure release, demonstrating good cyclic stability and repeatability. Stable cyclic performance is crucial for

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the reliability and durability of flexible pressure sensors in practical applications, indicating that the sensor can maintain stable performance during repeated use.

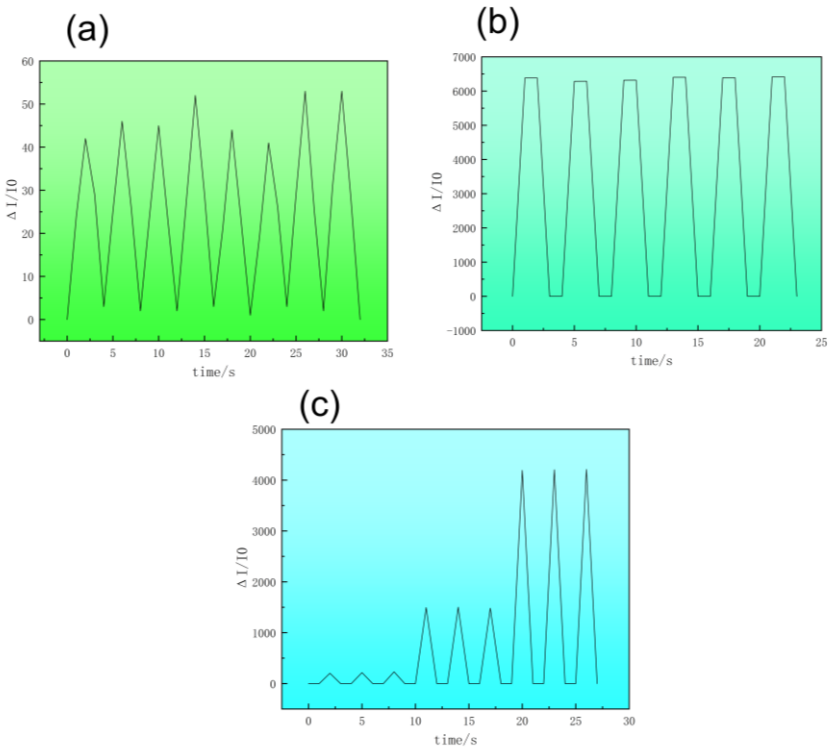

Figure 4: Simulated stepping, knee running (kicking legs), and breathing-these are the curves of the three applications

As shown in Figure 4, these are the curve figures of the sensor in three applications: simulated stepping, knee running (kicking), and breathing. In these three application scenarios—simulated stepping, knee running (kicking), and breathing—the flexible pressure sensor exhibits different response characteristics. During the simulated stepping process, the current variation rate ($\Delta I/I_0$) of the sensor shows a clear cyclical fluctuation, closely related to the rhythm of the stepping action. This cyclical change reflects the sensor's sensitive response to pressure changes during the stepping process, which is significant for gait analysis and motion monitoring. During the knee running (kicking) process, $\Delta I/I_0$ exhibits a more complex fluctuation pattern, related to the rhythm and force of the kicking action. This complex current variation indicates that the sensor can capture multiple pressure changes during the kicking action, which is significant for motion analysis and the field of sports science. During the breathing process, $\Delta I/I_0$ shows a relatively gentle fluctuation, related to the rhythm of breathing. This gentle current variation indicates that the sensor can accurately capture the minor pressure changes during the breathing process, which is significant for health monitoring and sleep research, demonstrating the potential applications of the prepared flexible pressure sensor in health monitoring, sleep research, and smart wearable devices.

3. Conclusion

This study successfully developed a flexible pressure sensor with a wide linear range and high sensitivity through the innovative design of a Janus dual-sensitive layer architecture. A biomimetic pyramid microstructure sensing layer was constructed using multi-level composite materials of

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CNT/CNF, combined with a PDMS stiffness modulation layer to achieve dynamic control of mechanical properties, solving the inherent contradiction between range-sensitivity and durability in traditional single-layer sensors. Experimental results showed that the sensor exhibited an outstanding sensitivity of 18.4 kPa-1 and an excellent linearity of 99.94% within a wide pressure range of 0–700 kPa, along with a rapid response time of 60 ms and a recovery time of 70 ms, demonstrating stable repeatability in cyclic loading tests. The Janus dual-sensitive layer achieved a phased switching of current transmission paths with pressure changes through the synergistic effect of the high-resistance layer and the low-resistance layer, effectively delaying the trend of resistance saturation and significantly widening the linear response range. The CNT/CNF composite sensitive layer not only optimized the dynamic response characteristics of the conductive network but also enhanced the mechanical properties of the material through nano-level dispersion effects, giving the sensor a 353.33% elongation at break and a tensile strength of 18 MPa. Furthermore, the sensor, while simplifying the fabrication process and reducing costs, showed broad application potential in fields such as intelligent robots, wearable health monitoring (e.g., gait analysis, respiratory detection), and human-computer interaction.

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